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Author(s): John S. Hoffman

Reviewed work(s):

Source: *Ambio*, Vol. 19, No. 6/7, CFCs and Stratospheric Ozone (Oct., 1990), pp. 329-333

Published by: [Springer](#) on behalf of [Royal Swedish Academy of Sciences](#)

Stable URL: <http://www.jstor.org/stable/4313728>

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Replacing CFCs: The Search for Alternatives

Chlorofluorocarbons will be replaced by other chemicals under the Montreal Protocol to Protect the Ozone Layer. While replacements have been found for virtually every use, they are not necessarily best in terms of greenhouse potential, energy efficiency, or other performance objectives. Additional possibilities have been identified, however, which could greatly reduce global warming and also provide superior performance. New working fluids and cycles for refrigerators coupled with advanced insulations could, for example, reduce global warming by more than one percent while saving consumers billions of dollars worldwide. Whether such advanced options come into play depends on how well industry and government can accelerate research and development and overcome institutional problems that could block acceptance.

INTRODUCTION

Chlorofluorocarbons (CFCs) have brought the world great economic benefits and serious environmental damage. Used in all types of refrigeration and air conditioning, as solvents, insulating and blowing agents, and in a variety of other goods and manufacturing processes, CFCs have depleted the global ozone column by 1.6% to 3% in the last eight years. As a consequence, the quantity of UV-B radiation over much of the earth's surface is 3–6% greater than what it would have been without ozone depletion. Over Antarctica every spring, the ozone layer is depleted by about 50%, yielding an increase in destructive UV-B radiation that exceeds 100%. The world responded to this threat with the Montreal Protocol, an international treaty which will eliminate CFC use by the year 2000 (1).

Substitutes have been identified for virtually every use of CFCs and reductions in a number of uses have already been accomplished by industry around the world (1). However, the challenge of finding replacements for CFCs has not been completed. Many of the original options may not be the best in terms of performance, cost, greenhouse forcing, toxicity, or other factors. Analysis and experiment demonstrate that better substitutes can significantly reduce society's energy consumption and its contribution to the greenhouse effect (1). Operating costs can be reduced as well. If these better alternatives prove workable and safe, they could reduce the costs of services such as cooling to levels below that attained with CFCs (1). Consequently, it is important that every option be carefully evaluated so that the best all-around selections can be made.

This task poses a substantial challenge to industry and governments worldwide. We must continue to explore and develop better alternatives as CFCs are phased out. Intensified investment in research and development and careful coordination between industry and government will be required to fully evaluate the performance

and environmental consequences of the various options. Even with such investment and coordination, industry could still commit to inferior substitutes unless industrial standards are quickly revised and contradictory environmental mandates are rationalized. Inevitably, tradeoffs will need to be considered between desirable goals, such as choosing between a minimal greenhouse impact and minimal flammability. The impacts on future welfare resulting from the many choices we will have to make over the next decade justify the strongest possible effort.

ORIGINS OF CFCs AND CONCERNS ABOUT THEM

CFCs were developed in the late 1920s as an answer to the prayers of the home refrigerator business. Prior to the 1930s, household refrigeration was either cumbersome (during ice deliveries), somewhat dangerous (where potentially hazardous refrigerants such as sulfur dioxide or methyl chloride were used), or non-existent. Mechanical home refrigerators were selling briskly, but safety lapses were causing serious problems (including death), and the future of the business was by no means secure. Thomas Midgely, a chemist with a General Motors associated research laboratory, was called in to find a safe refrigerant, and within a few days he settled on the CFC family as providing the best options. CFCs are compounds synthesized around one or two carbon atoms, with fluorine and chlorine atoms occupying all of the bonds with the carbon. CFC 12 emerged as an excellent candidate for home refrigerator use: stable, non-flammable, non-corrosive, and with good thermophysical attributes. Soon the DuPont Corporation (DuPont and General Motors were closely tied) put its technical capability and marketing skills behind CFCs. By the late 1940s, they had become the dominant refrigerant in refrigerators, helping

make widespread acceptance of household refrigeration possible (1).

With the CFCs established as profitable chemicals in one field, the chemical companies continued research and marketing efforts to find new uses for CFCs. In the 1950s, the aerosol industry was convinced to adopt them as propellants. Later, CFCs were used to create insulating foams. In the 1970s, CFC-113 was adopted as the primary cleaning agent for electronic components. By 1980, hundreds of products contained or were made using CFCs. CFC growth was truly phenomenal (Fig. 1).

Until 1971, there appeared to be no question that CFCs were safe, environmentally benign products; this was a key attribute in their successful marketing. In that year, however, a question never considered by Midgely was raised by James Lovelock, a British scientist: what happens to CFCs when they are released to the air? Using equipment made in his home, Lovelock found that CFC concentrations were essentially the same everywhere in the atmosphere (1). The implications were clear. CFCs must be accumulating in the atmosphere instead of raining out like most other chemicals. Subsequent measurements proved that atmospheric concentrations of CFCs were rising rapidly.

At about the same time, Harold Johnston and other atmospheric scientists were expressing concern that human activity could deplete the ozone layer. They hypothesized that a proposed fleet of supersonic airplanes could deposit large quantities of nitrogen oxides directly into the ozone layer, which would catalytically deplete this vital barrier to UV-B radiation (1). Similarly, Ralph Cicerone and Richard Stolarski were exploring another potential threat to the ozone layer; the proposed space shuttle was expected to transport chlorine to the stratosphere,

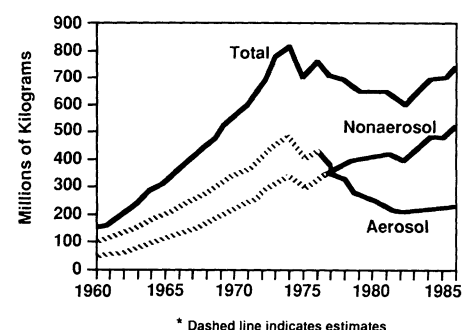


Figure 1. CFC-11 and CFC-12 production in the developed world. Estimates for the aerosol share of total production from 1960 to 1975 are based on the 1976 share reported in the CMA schedule (35).

which could also catalytically destroy ozone (1).

It turns out that neither supersonic airplanes nor space shuttles were required to carry ozone-destroyers into the stratosphere. F. Sherwood Rowland and Mario Molina recognized the connection between Midgely's discovery, Lovelock's finding and the ozone layer (1). Rowland and Molina hypothesized that because CFCs are virtually indestructible in the lower atmosphere, they would simply drift into the stratosphere, and would be broken down by high energy radiation, releasing ozone-depleting chlorine.

PUBLIC REACTION TO THE OZONE CRISIS

Public concern about the ozone layer grew quickly. By 1978, several nations had banned the use of CFCs in aerosols or capped production capacity levels. The chemical industry launched a search for non-chlorinated substitutes. Scientific research on stratospheric ozone was accelerated. From the first, it had been recognized that atmospheric models describing stratospheric ozone were simplified and incomplete, and that uncertainties existed

about the exact relationship between chlorine and ozone depletion. From year to year, estimates of depletion for any given level of chlorine changed as models were modified to reflect new information about the ozone layer's behavior (1). During this period, industry organized to prevent additional regulation, arguing that the scientific uncertainties were too great to justify additional regulation. Instead, they called for a system of assessment and review that would monitor the situation and respond when needed (1).

Research on substitutes was dropped as it became clear that there was no imminent threat of additional regulation (1). Scientific studies continued, however. In 1984, Michael Prather published a paper demonstrating that higher concentrations of CFCs had the potential to cause non-linear ozone depletion (1). Based on these results, workshops were held in 1985 on substitutes and their availability.

At those workshops it was revealed that research had been abandoned on the substitutes because they were more expensive than CFCs and therefore no business reason existed for pursuing them (12). This was the first time that the public had been told that substitutes were available,

and reaction to this statement was extremely negative (14, 10).

In 1985, the British Antarctic Survey announced the discovery of the ozone hole (10). The possibility that the ozone hole was natural prevented its acceptance as a basis for negotiations for controlling CFCs, (10) but the combination of growing environmentalism worldwide, models projecting future global ozone depletion, and the possibility that the ozone hole was caused by CFCs quickly led to increased public demand for protection of stratospheric ozone and intensified negotiations for CFC control.

In this political environment, hydrofluorocarbon 134a (HFC-134a) and hydrochlorofluorocarbon 123 (HCFC-123) were identified by several chemical companies as attractive alternatives to CFC-12 and -11 respectively. These chemicals were chosen because they had little or no chlorine, and they appeared to have promising thermophysical properties, similar to those of CFC-12 and -11. These compounds had undergone relatively little testing in end-use applications when they were selected as the options "most likely to succeed" in the summer of 1987 (17). Since 1987, it has become clear that the

CRITERIA FOR SELECTING REPLACEMENTS

A variety of environmental and economic criteria must be considered in selecting substitutes for CFCs.

- Their potential to contribute chlorine or other ozone depleting elements to the stratosphere.
- Their greenhouse effect (all CFC replacements are greenhouse gases, but they vary greatly in their potency).
- Their energy efficiency, which affects both their indirect contribution to greenhouse warming (via fuel combustion in power plants) and their operating costs.
- Their potential flammability, and consequent contribution to property and life losses from fires.
- Their potential toxicity and other detrimental effects on human health.
- Their potential for forming ground-based ozone.
- Their effectiveness in other respects, such as compatibility with materials or heat capacity.
- Their cost, including additional costs to overcome possible toxicity or safety concerns.

No compound is likely to be dominant in all criteria, so tradeoffs between objectives will be necessary. Table 1 shows some of the chemical substitutes identified to date and their characteristics for some of the above criteria. From the perspective of both the greenhouse effect and ozone layer depletion, HFC-152a, HCFC-124, HCFC-123, and HFC-32 emerge as attractive op-

tions, especially in high emitting uses. Much information still needs to be developed about all the options. Toxicity data are still incomplete. Flammability risks have not been reassessed. But it has become clear that differences

between options in terms of global impact on energy use, the greenhouse effect, and ozone depletion are significant. Under these circumstances, a complete analysis of options and tradeoffs is justified.

Table 1. Environmental attributes of CFCs and substitutes.

	Global warming potential (a)	Ozone depletion potential (b)	Commercial status	Status of toxicological testing (Feb. 1990)	Flammability
CFC-11	1500	1.0	in production	tested	none
CFC-12	4500	1.0	in production	tested	none
HFC-152a	47	0	in production	tested	mild
HCFC-123	29	0.013-0.027	in pilot production	in testing	none
HFC-32	(c)	0	was produced in limited quantities	limited testing	very mild
HCFC-124	150	0.013-0.030		in testing	none
HCFC-22	510	0.032-0.071	in production	tested	essentially none
HFC-134		0	in pilot production		none
HFC-134a	420	0	in pilot production	in testing	none
HCFC-142b	540	0.035-0.077	in production	tested	mild
HCFC-141b	150	0.065-0.14	in production	in testing	mild
HFC-125	860	0		preliminary testing	none
HFC-143a	1000	0		preliminary testing	?

(a) Obtained from Intergovernmental Panel on Climate Change (see reference 31). See also, WMO, NASA, and other sources (references 32-35). These GWP values are relative to carbon dioxide, which has been assigned a value of 1. These GWPs are assessed on a mass basis over a 500 year period. The three criteria used to assess GWPs, namely radiative forcing, atmospheric lifetime, and temporal integrating period, are still somewhat uncertain, so these data are subject to change. A shorter temporal span would increase the relative GWPs of HCFCs and HFCs.

(b) Data reflect the range of estimates from four different two-dimensional models. Data are relative to CFC-11, which has been assigned a value of 1. (see reference 33, Table IV).

(c) Not computed, but probably low.

equipment changes required to use HCFC-123 and HFC-134a are greater than expected, and that performance is worse. Other chemicals, including HCFC-124, HCFC-141b, HFC-152a, and HFC-32, along with chemical mixtures, have been identified as additional options.

Given past experience with the identification and subsequent use of CFCs we should be cautious in selecting substitutes. The short-term focus has proved disastrous in the past. Because the long-term environmental impacts of CFCs were ignored by Midgely and everyone else for 45 years, 3% of the world's ozone layer has been lost; an ozone hole over Antarctica will exist for at least a hundred years; and the greenhouse effect has been enhanced 25 percent. It also appears that, on the basis of preliminary testing, energy costs have been higher than necessary, because CFC-12 was chosen over more energy-efficient compounds such as HFC-152a, which were ignored or rejected (18).

Thus, in deciding on future substitutes, it is critical that careful testing be done on many options, and that all of their advantages and disadvantages be carefully evaluated. It is also critical to consider the better technology available to use these chemicals. CFC-11 and -12 were chosen to remedy problems that existed with leaky 1930s refrigeration technologies. The search for the best alternatives should evaluate options based on the conditions found in technologies that exist today or could emerge in the 1990s and beyond.

REPLACEMENTS UNDER CONSIDERATION AND THEIR POTENTIAL IMPACTS

Solvent Uses

A variety of non-chlorine based substitutes offer attractive replacements for CFCs in electronics cleaning. CFC-113 has been the solvent of first choice because of strong technical support, prescriptive military specifications, and its generally good performance. Until recently, exploration of other options had been limited. However, the ozone crisis has catalyzed a very strong user-led effort with strong government-industry cooperation. This effort has already succeeded in finding a large number of terpenes, alcohols, and aqueous-based alternatives which appear competitive in terms of both cost and performance and pose essentially no greenhouse threat (19). The toxicity, flammability, and disposal of some of these substitutes are still being investigated, but it appears that many viable and probably superior options will emerge to replace CFC-113.

Aerosols

There are many practical and cost-effective alternatives for aerosol applications. Where ground-based ozone is not a problem, hydrocarbons can provide less expensive and equally effective propellants (10). Dimethylether is an option for polluted regions (10). In addition, there are a variety of other alternative products such as sprays, pumps, and roll-ons that provide good service at lower cost to the consumer (21).

One large unanswered question raised by the CFC phaseout is the future of HCFC-22. HCFC-22 has a somewhat shorter lifetime than CFC-12 or -11 because one of the four bonds to its carbon atom is occupied by a hydrogen atom rather than by a fluorine or chlorine atom. This causes much of the HCFC-22 to be oxidized in the lower atmosphere before it reaches the stratosphere. A given quantity of HCFC-22 emitted to the atmosphere is reduced by approximately two-thirds in 15 to 20 years after release (the 2/3 reduction target is the convention used for "lifetime" in these analyses). Thus its ozone depletion potential is reduced to five percent of CFC-11's.

HCFC-22 is already widely used in heat pumps and home air conditioners. However, HCFC-22 could be used in virtually any refrigeration task (commercial and office refrigeration are two good examples) if the equipment were redesigned, although it would incur an energy penalty in some uses.

Generally, many of the participants in the Montreal Protocol have viewed HCFC-22 as a bridging alternative to be replaced in 40 to 60 years as better options develop. Nevertheless, some users have expressed reservations about using HCFC-22 in high emission applications over the next thirty years, because of its high level of chlorine and

relatively strong greenhouse warming potential. To fully examine the greenhouse effect of this compound, however, one must also consider its energy consumption during use. Used in refrigerator mixtures, for example, in which higher efficiency was obtained, it might be the case that reduced CO₂ emissions would greatly outweigh the direct warming produced by HCFC-22 leakage. Used in supermarket freezer cases with high emissions, the reverse might be the case (4).

HFC-32 in mixtures with other compounds probably represents a strong alternative to HCFC-22 in many uses (29). HFC-32 has a good coefficient of performance, a much shorter lifetime, no chlorine, has higher heat capacity, and is only very mildly flammable. Some mixture containing HFC-32 may operate as efficiently or even more efficiently than HCFC-22 alone, and may have no flammability problem whatsoever. Testing on this compound in various applications is still in the planning stages. HFC-32 was produced in small quantities in the 1960s, and the somewhat limited toxicological tests performed at the time indicated no serious problems when HFC-32 was adequately separated from HCFC-31 (which is co-produced in its synthesis) (30).

Refrigerants in Household Refrigerators

Refrigerators emit small amounts of CFC-12, usually at the end of the product's life. All of the options to replace CFC-12 have relatively low ozone depletion potentials and direct emission would contribute negligibly to global warming. Refrigerators also consume large amounts of electricity, making operating costs a significant factor, especially given their widespread use. Because most electricity is generated with fossil fuels, the greenhouse impact associated with the CO₂ emissions from this electricity generation can be quite significant. Thus, for refrigerators, the energy-efficiency of proposed alternatives can become a key criterion while the direct greenhouse effect and ozone depletion potentials of the substitutes may be of lesser importance.

HFC-134a was an early favourite to replace CFC-12 and is still under active consideration (10). However, tests of HFC-134a indicate that it will increase energy consumption by 8–15% in current refrigerators and perhaps by 0–5% if the compressors were improved (22). Any technical improvements in compressors used to compensate for deficiencies in HFC-134a's intrinsic performance would use up energy efficiency gains that would have otherwise been available.

Other options—potentially much more attractive from an energy standpoint—still await complete evaluation. HFC-152a has been tested in a few refrigerators with efficiency improvements in the 3–7% range, which would significantly reduce energy

costs and greenhouse-gas emissions compared with HFC-134a (18, 22). In the United States the ten-year cost savings of this refrigerant (once in place for all refrigerators) would be approximately USD 861 million with large reductions in CO₂ emissions (4). HFC-152a has been used in mixtures in dehumidifiers and truck transport, where it has proved to be compatible with existing equipment, something that HFC-134a has yet to do.

HFC-152a may have a different problem: flammability. Investigations are now underway to assess the fire risk associated with HFC-152a use in refrigerators. Initial results are promising since HFC-152a appears more difficult to ignite than refrigerants like propane and would be used in small quantities (3–4 ounces) (10). Furthermore, compared with risks already existing in kitchens, such as natural gas stoves and larger quantities of more flammable hydrocarbons in aerosols, the relative risks from HFC-152a appear small.

In the United States, manufacturers have publicly stated that their major concern with HFC-152a is not with its real risks, but with the potential lawsuits that they believe switching to a flammable refrigerant could entail (10). The US EPA and Underwriters Laboratory are conducting a public process aimed at assessing all the real risks from refrigerants, giving the companies a defensible basis for using HFC-152a if it proves as safe as preliminary analyses indicate.

Certain refrigerant mixtures, such as

142b/22 and 152a/22/124 are also options for refrigerators. While both contain flammable components, the mixtures themselves are inflammable. Both could be expected to improve energy performance, although less than HFC-152a alone would. The three-component blend would likely require less of a change in refrigerator design (22). Although both would contribute some chlorine to the atmosphere, the quantities are small.

Finally, several non-azeotropic mixtures have produced some very promising results in experiments conducted by EPA contractors. These mixtures have components with boiling points that are widely separated and would be used in dual

evaporator refrigerators with two intercoolers, patterned after a design developed by A. Lorenz in the 1970s (See Fig. 2). Lorenz achieved energy efficiency gains of 20 percent, and model calculations indicate such improvements are possible with several mixtures (25, 10).

Refrigerators in much of the world already use two evaporators (but not the second intercooler), so modifying their design to the Lorenz pattern would be a comparatively modest job. In countries like the US, where single evaporator models are used, the required changes would be greater, as would the environmental, energy, and cost benefits. Fully implemented on a global basis the energy savings com-

pared with CFC-12 could exceed USD 10.5 billion per year (3). Other options—such as dual loop systems with two compressor/evaporator/condenser systems using HFC-152a and 142b or other refrigerants—could yield similar energy and cost savings (R. Merriam, Arthur D. Little, Inc., pers. comm.).

Refrigerator Insulation

HCFC-123 and HCFC-141b are both potential replacements for CFC-11 in the insulation of refrigerators. Current tests indicate that these replacements would have slightly worse energy performance (5%), which could be made up by slightly thickening the walls of the refrigerator (26). A much better alternative—vacuum insulation—could provide energy improvements of 20% or more (3). One cost-effective, long-lived vacuum technology has already been applied to uses such as district heating pipes (10). Adapting this technology to refrigerators would require significant retooling. Other options are also being pursued, including aerogels and particle vacuums. While identifying the best technology awaits further testing, it is clear that economically and environmentally superior products can be developed. One additional barrier might stand in the way of using vacuum insulation in household refrigerators: higher first cost. However, as energy efficiency and the greenhouse effect emerge as more serious issues, rebates, alternative financing, and governmental regulation are likely to solve this problem.

Automobile Air Conditioning

HFC-134a is clearly the leading option to replace CFC-12 in automobile air-conditioners. Automobile air-conditioners differ from refrigerators in that their energy use is relatively low, but refrigerant emissions are quite high. HFC-134a would still contribute significantly to greenhouse warming due to its relatively long lifetime (20 years) and its significant greenhouse potential. Using HFC-134a in mobile air-conditioners would add a 0.34% increment to the global warming expected by 2075 (3). HFC-134a would pose no threat to the ozone layer because it contains no chlorine.

HFC-152a has not been seriously considered in automobiles due to its perceived flammability risk. It has a direct greenhouse effect 90% lower than HFC-134a's. Further cost and energy savings would also accrue due to higher energy efficiency and reduced weight. No serious review of the actual fire risks from using HFC-152a has been conducted yet. HFC-152a contains no chlorine, so it would pose no risk to the ozone layer.

The binary mixture of 142b/22 has been tested in automobiles and could provide adequate service. However, HCFC-142b is flammable and consequently provides no advantage over HFC-152a in that respect. It would also have a larger direct greenhouse effect than HFC-134a, and a larger ozone depletion potential as well. The ternary mixture of 152a/22/124 would not pose the same fire risk as 142b/22 or HFC-152a alone, because of differences in their leakage properties, but would probably

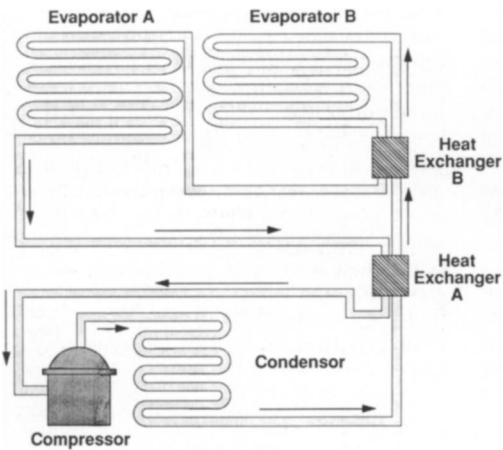


Figure 2. Lorenz cycle refrigerator/freezer. Lorenz cycles are predicted to increase energy efficiency by using non-azeotropic mixtures. A non-azeotropic mixture uses refrigerants with widely separated boiling points. Because different cooling requirements exist in the freezer and refrigerator (provisions) part of refrigerator/freezers, this approach has the potential to improve performance (25).

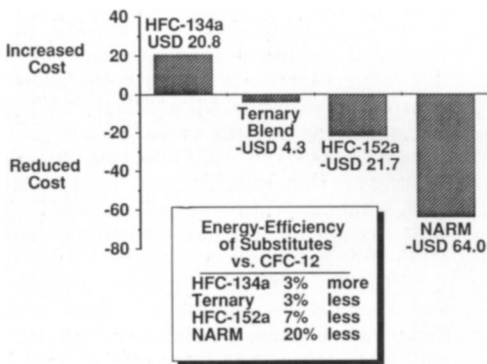
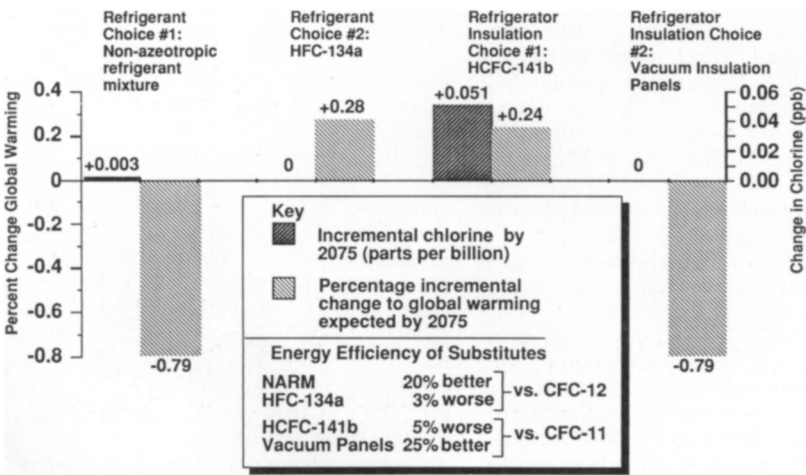


Figure 3. Incremental social costs and benefits of choosing refrigerants for household refrigerators (US only; cumulative 1989 to 2075; USD in 1985 billions) (4).

Figure 4. Global environmental impacts of choosing household refrigeration technology (3).



have greater emission rates than HFC-134a, thereby making it unattractive for new cars but potentially attractive for retrofits (28).

Other options, including compressed air, helium systems, and hermetically sealed systems have also been discussed. None have received significant consideration for the short term. All would offer major environmental benefits if proven practical, but would take longer to develop than modifying current systems. Given the prospective deadline for phaseout, it is probably necessary that HFC-134a be used in the first generation of cars. Other options may prove more desirable in the longer run.

Chillers

Chillers (refrigerating devices that cool large buildings) use several different CFCs, depending on the size and design of the machine. The largest uses are for CFC-11 in centrifugal chillers and HCFC-22 in reciprocating models; CFC-12 and R-500 (a mixture of 23% 152a/77% 12) are also used in some applications. At this time HCFC-123 appears to be the best option for centrifugal chillers, although it has a lower heat capacity. New installations in this market could also be serviced with ammonia systems (if safety concerns can be overcome), probably at energy savings of several percent (3). Lithium bromide systems may also offer significant energy savings.

Other Uses

There are dozens of other products and processes that use CFCs, including dry cleaning, sterilizing medical instruments, cold storage, and truck transport. Options to replace CFC-11, -12, and -113 have been found for all these uses (2). It may be that a more extensive search for replacements would yield still better options.

CONCLUSION

Given the importance of phasing out the fully halogenated CFCs and the timeframe likely to emerge for that to happen (2000), it is natural that many producers and users of CFCs want to decide on replacements now. The long lifetime of equipment, retooling many product lines, and limited capital all suggest that sooner is better than later. However, rushing forward too quickly would be taking a grave risk. EPA has conducted several analyses which demonstrate the overall economic and environmental importance of finding the best substitutes. Figure 3 shows the cost differences between using HFC-134a or a better alternative as a substitute for CFC-12 in household refrigerators. Figure 4 shows the difference in the greenhouse impact and stratospheric chlorine loading as a result of choosing refrigerant and foam technology. These are only some of the differences that could emerge from selection of different options.

Given these important differences, it is imperative that governments and industry throughout the world invest heavily in accelerating the search for the best substitutes. The investigation must encompass all factors: energy, ozone depletion poten-

tial, greenhouse warming potential, toxicity, flammability, performance, and cost.

The selection of options will inevitably require tradeoffs. To date, too little effort has been invested in considering the full range of options compared to the potential value that preliminary analysis indicates some choices may have. History shows that once made, choices will have a life of their own, but that inevitably choices with

bad effects will prove unacceptable and necessitate change. Neither government nor industry alone can develop the best options. The time left and the social choices required necessitate strong cooperation and intense investment. Rather than repeat history, we should learn from it. By evaluating the alternatives for all critical factors we can select the best ones for the future.

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37. Acknowledgment: Robert Kwartin, David De-Busk, and William Wechsler provided valuable research and editing support. Eileen Claussen and Steve Seidel of USEPA and anonymous reviewers provided many helpful comments. My thanks to all of them.

John S. Hoffman is Director of the Global Change Division, Air Office, United States Environmental Protection Agency. During his tenure he has authored numerous articles on the greenhouse effect, served as senior author of EPA's Risk Assessment on Stratospheric Ozone, directed projects to find improved substitutes for CFCs, and provided technical support to negotiations that led to the Montreal Protocol. His address: EPA Global Change Division, ANR-445, 401 M Street, SW, Washington DC 20460, USA.